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The relationship between the crystallization process and the soft magnetic properties of nanocrystalline Fe–M–B–Cu (M=Zr, Nb) alloy

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The relationship between the crystallization process and the soft magnetic properties of nanocrystalline $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ alloy has been studied by comparison with that of $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ alloy. When the annealing temperature T_a is slightly above the crystallization temperature, high permeability can only be obtained for Fe–Nb–Zr–B–Cu after annealing for very short times. The T_a dependence of the coercive force of Fe–Nb–Zr–B–Cu cannot be explained by the change of the grain size of the bcc phase. The soft magnetic properties of Fe–Nb–Zr–B–Cu is dominated by not only the grain size but also the Curie temperature of the intergranular amorphous phase. It is concluded that the magnetic softness of Fe–Nb–Zr–B–Cu is related directly to the degree of the reduction in the apparent anisotropy, while that of Fe–Si–B–Nb–Cu is strongly affected by the Si content of the bcc phase. [S0021-8979(97)25208-4]

I. INTRODUCTION

Recently, nanocrystalline soft magnetic alloys consisting of bcc nanoscale crystallites embedded in a residual amorphous phase have been obtained by crystallizing melt-spun amorphous ribbons.[1–7] It has been found that Fe based alloys containing a small amount of Cu such as Fe–Si–B–Nb–Cu[1,2] and Fe–Nb–Zr–B–Cu[4–7] exhibit high effective permeability μ_e above 1×10^5 at 1 kHz. In particular, the nanocrystalline Fe rich Fe–Nb–Zr–B–Cu alloys are attractive because the alloys exhibit high μ_e of 1.0×10^5–1.6×10^5 at 1 kHz and a high saturation magnetic flux density B_s of 1.5–1.6 T, simultaneously.[4–7] In this work, we have investigated the relationship between the crystallization process (annealing time and temperature) and the soft magnetic properties of a nanocrystalline $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ alloy by comparison with that of the $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ alloy.

II. EXPERIMENT

A single roller melt spinning method in an Ar atmosphere was used to produce the rapidly solidified ribbons with 15 mm width and about 20 μm thickness. Annealing treatment of the as-quenched samples was carried out by treating the samples for 0–3.6 ks at various temperatures in a vacuum; the heating rate was 0.67 K/s. The crystallization temperature of the amorphous alloys was determined by a differential scanning calorimeter (DSC) using the same heating rate as in the annealing treatment. The mean grain size D_{bcc} was evaluated from the half-width of the bcc (110) x-ray diffraction peak. The saturation magnetic flux density B_s and the coercivity H_c were measured with a vibrating sample magnetometer (VSM) and a low frequency B-H loop tracer, respectively. The effective permeability μ_e under an applied field of 0.4 A/m was measured with a vector impedance analyzer. The saturation magnetostriction λ_s was measured by using a strain gage technique.

III. RESULTS AND DISCUSSION

Figure 1 shows the annealing time t_a dependence of (a) B_s, (b) μ_e, (c) λ_s, (d) D_{bcc}, and (e) the lattice parameter a_{bcc} of the bcc phase for the $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ (closed circles) and $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ (open circles) alloys when the annealing temperature T_a is slightly above the crystallization temperature T_x. The crystallization temperature determined by DSC is 781 K for Fe–Nb–Zr–B–Cu and 821 K for Fe–Si–B–Nb–Cu. The annealing temperatures was chosen as T_x+2 K. The saturation magnetic flux density of both alloys is independent of t_a except for Fe–Nb–Zr–B–Cu at $t_a=0$ s. (Here, $t_a=0$ s means that the sample was heated up to T_a first at a heating rate of 0.67 K/s, and it was cooled at a cooling rate of -0.67 K/s as soon as the temperature reached T_a.) A high μ_e value above 8×10^4 has been obtained for Fe–Nb–Zr–B–Cu. The μ_e and D_{bcc} values of Fe–Nb–Zr–B–Cu are almost constant ($\mu_e=7.3\times10^4$–8.3×10^4, $D_{\mathrm{bcc}}\approx7.5$ nm) in the t_a range of 0 to 3.6 ks. On the other hand, μ_e of Fe–Si–B–Nb–Cu significantly decreases with a decrease of t_a from $\mu_e=7.5\times10^4$ at $t_a=1.8$ ks to $\mu_e=3.5\times10^4$ in $t_a\leqslant60$ s though D_{bcc} gradually decreases.

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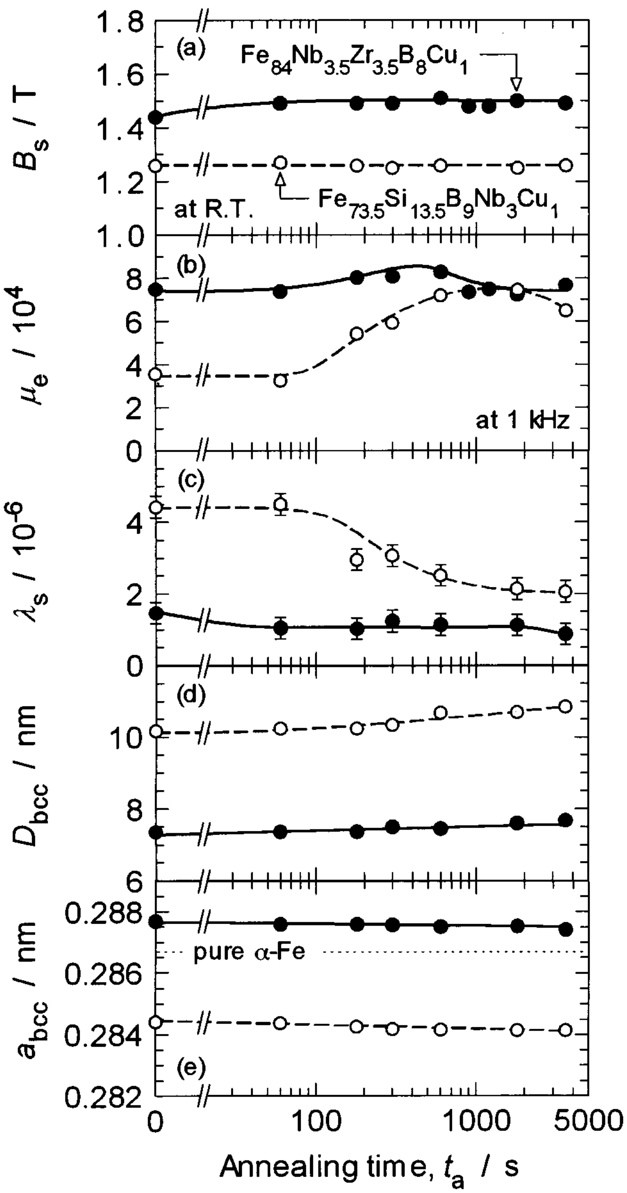

FIG. 1. Annealing time dependence of (a) B_s, (b) μ_e, (c) λ_s, (d) $D_{\rm bcc}$, and (e) $a_{\rm bcc}$ for nanocrystalline $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ (annealed at 783 K) and $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$ (annealed at 823 K) alloys. The annealing temperatures were chosen as T_x+2 K.

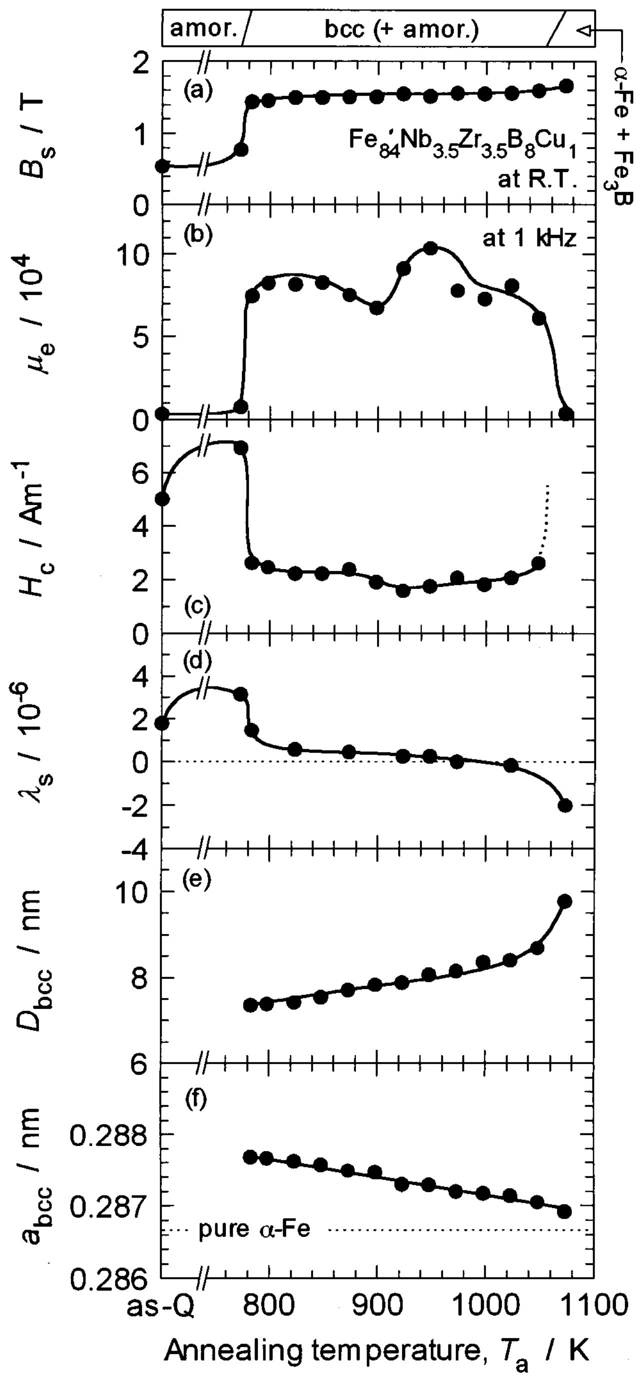

FIG. 2. Annealing temperature dependence of (a) B_s, (b) μ_e, (c) H_c, (d) λ_s, (e) $D_{\rm bcc}$, and (f) $a_{\rm bcc}$ for the $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ alloy. The samples were heated up to T_a at a heating rate of 0.67 K/s, and they were cooled as soon as the temperature reached T_a.

The magnetostriction of both alloys shows positive values and gradually increases with decreasing t_a. However, the t_a dependence of λ_s for Fe–Si–B–Nb–Cu is considerably larger than that of Fe–Nb–Zr–B–Cu. The lattice parameter of the bcc phase is almost independent of t_a for Fe–Nb–Zr–B–Cu. On the other hand, for Fe–Si–B–Nb–Cu, the lattice parameter slightly increases with decreasing t_a. These results suggest that μ_e of the Fe–Nb–Zr–B–Cu alloy is mainly dominated by the grain size, whereas μ_e of Fe–Si–B–Nb–Cu may be strongly affected by other factors. It has been reported for Fe–Si–B–Nb–Cu that the Si content of the bcc phase (which depends strongly on the annealing time[8]) plays an important role in obtaining high μ_e through the reduction in λ_s.[2]

Figure 2 shows the T_a dependence of (a) B_s, (b) μ_e, (c) H_c, (d) λ_s, (e) $D_{\rm bcc}$, and (f) $a_{\rm bcc}$ of Fe–Nb–Zr–B–Cu. The samples were heated up to T_a at a heating rate of 0.67 K/s, and they were cooled as soon as the temperature reached T_a. The structures of the samples examined by x-ray diffractometry are also shown. It has been reported that for Fe–Si–B–Nb–Cu, good soft magnetic properties can be obtained after crystallizing only in a T_a range of about 770 to 850 K (when t_a=3.6 ks).[2] On the other hand, for Fe–Nb–Zr–B–Cu, good soft magnetic properties were ob-

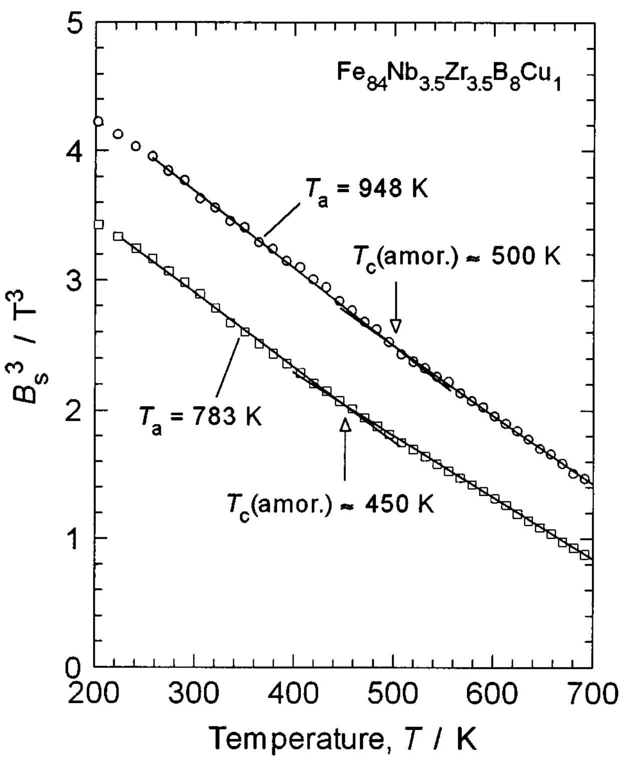

FIG. 3. Temperature dependence of cubed saturation magnetic flux density for $Fe_{84}Nb_{3.5}Zr_{3.5}B_8Cu_1$ alloys heated up to 783 and 1048 K.

tained after crystallizing in a wide T_a range of about 780 to 1050 K. With increasing T_a, B_s rises abruptly at around $T_a \approx 780$ K due to a structural change from the amorphous phase to the nanocrystalline bcc phase, and then gradually increases. The magnetostriction shows the rather large positive value of 2×10^{-6}–3×10^{-6} at the amorphous state, falls abruptly due to the structural change, and then changes to a negative value of -2×10^{-6} at $T_a=1073$ K passing through zero with increasing T_a. The change of λ_s with T_a is presumably due to a changing amount of solute elements in the bcc phase. The lattice parameter is slightly larger than that of pure Fe, indicating that the bcc phase has dissolved some solute elements. The lattice parameter decreases with increasing T_a, reflecting the decrease in the amount of solute elements in the bcc phase. As shown in Fig. 2(b), μ_e exhibits two maxima around $T_a \approx 820$ K and around $T_a \approx 950$ K. The alloy annealed at 948 K exhibits a high μ_e above 1×10^5, a high B_s of 1.52 T, and a sufficiently small λ_s of $+0.3\times10^{-6}$, simultaneously.

The origin of the excellent soft magnetic properties for the nanocrystalline alloys has been explained on the basis of the random anisotropy model,[2] which states that the magnetocrystalline anisotropy is averaged out due to strong ferromagnetic exchange coupling between the ferromagnetic grains. According to this model, the soft magnetic properties are improved as D_{bcc} becomes smaller, e.g., H_c is proportional to $D_{bcc}{}^6$. However, as shown in Fig. 2(c), H_c exhibits a minimum around $T_a \approx 920$ K though D_{bcc} increases with increasing T_a. This behavior cannot be explained by the T_a dependence of D_{bcc}.

In Fe–Nb–Zr–B–Cu, the intergranular exchange coupling is mediated by a residual amorphous phase surrounding the bcc grains.[4] If the Curie temperature T_c(amorphous) of the residual amorphous phase is low, it cannot fully mediate the intergranular exchange coupling at room temperature due to the thermal fluctuation of the spins in the residual amorphous phase. Figure 3 shows a B_s^3 versus temperature plot for Fe–Nb–Zr–B–Cu heated up to 798 and 948 K. The Curie temperature of the residual amorphous phase was evaluated from the inflection points of the B_s^3-T curves. It is difficult to determine T_c(amorphous) exactly because the change of B_s due to the ferromagnetic to paramagnetic phase transition of the residual amorphous phase is very small. However, it can be said that with increasing T_a, the T_c(amorphous) increases and the exchange coupling between the bcc grains is enhanced. In the lower T_a range, H_c gradually decreases with increasing T_a because the intergrain coupling is enhanced. On the other hand, in the higher T_a range, the deterioration of magnetic softness by the increase in D_{bcc} probably becomes dominant. It is concluded that the magnetic softness of Fe–Nb–Zr–B–Cu relates directly to the degree of the reduction in the apparent anisotropy, while that of Fe–Si–B–Nb–Cu is strongly affected by the Si content of the bcc phase.

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